

FORM PTO-1390
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U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY DOCKET NUMBER

AM100246-00

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

U.S. APPLICATION NO (IF KNOWN SEE 37 CFR 1.5)

10/019481

INTERNATIONAL APPLICATION NO.

PCT/US 00/17895

INTERNATIONAL FILING DATE

June 28, 2000

PRIORITY DATE CLAIMED

July 5, 1998

TITLE OF INVENTION: **ANT Controllers and Method for Application Thereof**

APPLICATION(S) FOR DO/EO/US: **Takagi, Kazuhiro, et al.**

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include the items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ Has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ An English language translation of the International Application as filed (35 U.S.C. 371(C)(2)).
 - a. ☐ is attached hereto.
 - b. ☐ Has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☒ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made
8. ☒ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 20 below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included
13. ☐ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A Change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☐ Other items or information:

U.S. APPLICATION NO. (If known see 37 C.F.R. 1.5) 10/019481		INTERNATIONAL APPLICATION NO. PCT/US00/17895		ATTORNEY'S DOCKET NUMBER AM100246-00	
21. <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS PTO USE ONLY	
Basic National Fee (37 CFR 1.492(a)(1)-(5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1040.00					
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$890.00					
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$740.00					
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)..... \$710.00					
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$740.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
CLAIMS	Number Filed	Number Extra	Rate		
Total claims	19 - 20 =	0	X \$18.00	\$0.00	
Independent claims	1 - 03 =	0	X \$80.00	\$0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$270.00	
TOTAL OF ABOVE CALCULATION =				\$0.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$	
SUBTOTAL =				\$1010.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$1010.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$40.00	
TOTAL FEES ENCLOSED =				\$0.00	
				Amount to be:	
				Refunded	
				Charged	
				\$1,050.00	

a. ☐ A check in the amount of \$_____ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 02-1197 in the amount of \$1050.00 to cover the above fees
A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 02-1197. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be changed to a credit card. **WARNING:** Information on this form may become public. **Credit card** Information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:
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31,278
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JC13 Rec'd PCT/PTO 31 DEC 2001

ANT CONTROLLERS AND METHOD FOR
APPLICATION THEREOF

5

BACKGROUND OF THE INVENTION

FIELD OF THE INVENTION

The present invention relates to novel ant
controller containing a hydrazine derivative as an
10 active ingredient and to a method for application of the
ant controller.

RELATED ART

The hydrazine derivatives represented by the
15 formula (I) which can be used as active ingredient of
the ant controllers of the present invention are known
compounds disclosed in JP-A-5-4958, JP-A-5-17428, JP-A-
5-32603, JP-A-5-262712, etc. In these patents, it is
described that these derivatives have an insecticidal
20 activity as agrihorticultuarl insecticides against
LEPIDOPTERA such as diamondback moth, rice leafroller,
etc., HEMIPTERA such as tea green leafhopper, pear lace
bug, etc., COLEOPTERA such as twenty-eight-spotted
ladybird, maize weevil, etc., DIPTERA such as melon fly,
25 house fly, house mosquito, etc., and TYLENCHIDA such as
coffee root-lesion nematode, root-knot nematode, etc.

Any of these patent gazettes, however, does
neither describe nor suggest that said hydrazine
derivatives have a marked insecticidal effect against

"TELETYPE" 040404

5 ISOPTERA such as formosan subterranean termite, kolbe, etc., HYMENOPTERA such as cabbage sawfly, Carpenter ant, etc., ORTHOPTERA such as Japanese cockroach, field cricket, rice grasshopper, etc., and PSOCOPTERA such as large pale booklouse, etc.

10 SUMMARY OF THE INVENTION

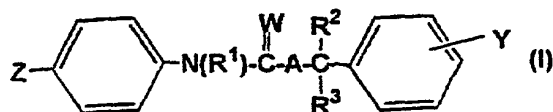
The present inventors have conducted extensive studies with the aim of creating a novel ant controller having a marked controlling effect upon ants doing harm to the wooden materials constituting houses, furniture, etc. or crops and human being. As a result, it has been found that some of the hydrazine derivatives described in the above-mentioned prior art have a marked insecticidal effect upon termites and ants. The present invention has been accomplished on the basis of this

20 findings.

DETAILED DESCRIPTION OF THE INVENTION

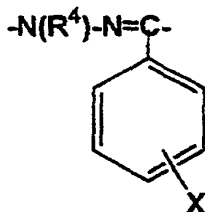
The present invention relates to ant controllers containing as active ingredient thereof a hydrazine derivative represented by the following

25 formula (I) and method for application of the ant controllers:

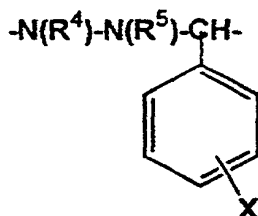


wherein A represents:

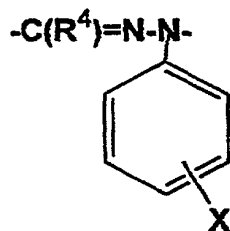
- 3 -



- 5 (wherein R^4 represents hydrogen atom or C_1-C_6 alkyl group, and X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C_1-C_6 alkyl group and halo C_1-C_6 alkyl group),

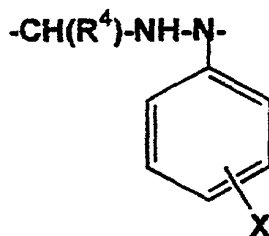


- 10 (wherein R^4 and X are as defined above, and R^5 represents hydrogen atom, C_1-C_6 alkylcarbonyl group or phenylcarbonyl group which may have 1 to 2, same or different substituents selected from the group consisting of C_1-C_6 alkyl groups),
- 15 alkyl groups),



(wherein R^4 and X are as defined above), or

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(wherein R^4 and X are as defined above);

5 R^1 represents hydrogen atom or C_1-C_6 alkyl group;

R^2 and R^3 , which may be same or different, represent hydrogen atom, hydroxyl group, C_1-C_6 alkyl group, C_1-C_6 alkoxy group, C_1-C_6 alkylcarbonyl group or
10 phenylcarbonyl group;

Y represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, nitro group and cyano group;

15 Z represents halogen atom, cyano group, C_1-C_6 alkyl group, halo C_1-C_6 alkyl group, C_1-C_6 alkoxy group, halo C_1-C_6 alkoxy group, halo C_1-C_6 alkylthio group, halo C_1-C_6 alkylsulfinyl group or halo C_1-C_6 alkylsulfonyl group; and

20 W represents oxygen atom or sulfur atom.

The ant controller of the present invention is an excellent ant controller for protecting wooden materials such as trees, board fences, sleepers, etc. and buildings such as shrines, temples, houses, outhouses,
25 factories, etc. from ants such as termites, and for controlling ants doing harm to crops or human being.

5 In the definition of the formula (I) shown
above, the term "halogen atom" means chlorine atom,
bromine atom, iodine atom and fluorine atom; the term
"C₁-C₆ alkyl" means a straight or branched chain alkyl
group having 1 to 6 carbon atoms; and the term "halo C₁-
10 C₆ alkyl" means an alkyl group having 1 to 6 carbon atoms
substituted with at least one, same or different halogen
atoms.

Preferable examples of the hydrazine
derivative represented by the formula (I) of the present
15 invention are the hydrazine derivatives represented by
the formulas (I-1) and (I-2) as mentioned below.
Preferable examples of each substituent of the hydrazine
derivatives of formulas (I-1) and (I-2) are the
compounds wherein W is oxygen atom, X is trifluoromethyl
20 group, Y is cyano group, Z is trifluoromethoxy group,
and each of R¹, R², R³ and R⁴ is simultaneously a hydrogen
atom. More preferable examples are the compounds
wherein X is substituted on the 3-position, and Y is
substituted on the 4-position of the phenyl ring.

25 Most preferable example is the hydrazine
derivative represented by the formula (I-1), wherein
each of R¹, R², R³ and R⁴ is simultaneously a hydrogen
atom, X is trifluoromethyl group substituted on the 3-
position of the phenyl ring, Y is cyano group
30 substituted on the 4-position of the phenyl ring, and Z
is trifluoromethoxy group.

Typical examples of the hydrazine derivative

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5 represented by the formula (I) used as an active ingredient of the ant controller of the present invention are shown in Table 1 to Table 4, but the present invention is by no means limited to the compounds exemplified herein.

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Formula (I-1)

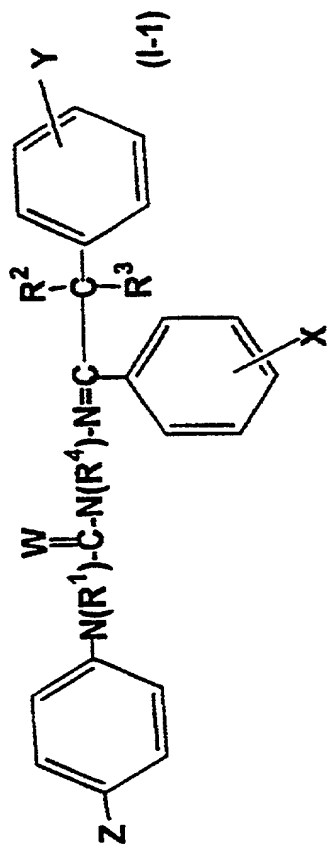


Table 1

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp (l)
1	H	H	H	H	H	H	Cl	O	199
2	H	H	H	H	H	H	OCF ₃	O	149
3	H	H	H	H	H	4-Cl	Cl	O	206
4	H	H	H	H	H	4-Cl	OCF ₃	O	197
5	H	H	H	H	H	4-CN	Cl	O	217
6	H	H	H	H	H	4-CN	Cl	S	128
7	H	H	H	H	H	4-CN	OCF ₃	S	116

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp °C
8	H	H	H	H	H	4-CN	OCF ₃	O	214 E-form
9	H	H	H	H	H	4-CN	OCF ₃	O	159 Z-form
10	H	H	H	H	H	4-NO ₂	Cl	O	222
11	H	H	H	H	H	4-NO ₂	Cl	S	206
12	H	H	H	H	H	4-NO ₂	OCF ₃	O	189
13	H	H	H	H	H	4-NO ₂	OCF ₃	S	139
14	H	H	H	H	H	4-NO ₂	SCF ₃	O	200
15	H	H	H	H	3-Cl	H	OCF ₃	O	212
16	H	H	H	H	3-Cl	4-Cl	OCF ₃	O	201
17	H	H	H	H	3-Cl	4-CN	Cl	O	206

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp °C
18	H	H	H	H	3-Cl	4-CN	OCF ₃	0	187 E-form
19	H	H	H	H	3-Cl	4-CN	OCF ₃	0	148 Z-form
20	H	H	H	H	3-Cl	4-CN	OCF ₃	S	199
21	H	H	H	H	3-Cl	4-CN	SCF ₃	0	215
22	H	H	H	H	3-Cl	4-CN	SOCF ₃	0	205
23	H	H	H	H	3-Cl	4-CN	SO ₂ CF ₃	0	212
24	H	H	H	H	3-Br	H	Cl	0	191
25	H	H	H	H	3-Br	H	OCF ₃	0	209
26	H	H	H	H	3-Br	4-CN	Cl	0	205
27	H	H	H	H	3-Br	4-CN	OCF ₃	0	176
28	H	H	H	H	3-Br	4-CN	SCF ₃	0	206

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp °
29	H	H	H	H	3-Br	4-CN	SO ₂ CF ₃	0	216
30	H	H	H	H	3-Br	4-CN	SO ₂ CF ₃	0	215
31	H	H	H	H	3-F	H	Cl	0	206
32	H	H	H	H	3-F	H	OCF ₃	0	200
33	H	H	H	H	3-F	4-Cl	OCF ₃	0	191
34	H	H	H	H	3-F	4-Cl	Cl	0	208
35	H	H	H	H	3-F	4-CN	OCF ₃	0	202
36	H	H	H	H	3-I	4-CN	Cl	0	213
37	H	H	H	H	3-I	4-CN	OCF ₃	0	201
38	H	H	H	H	3-CH ₃	H	Cl	0	185
39	H	H	H	H	3-CH ₃	H	OCF ₃	0	198
40	H	H	H	H	3-CH ₃	4-CN	Cl	0	200
41	H	H	H	H	3-CH ₃	4-CN	OCF ₃	0	189

[illegible][illegible]

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp [°C]
53	H	CH ₃	H	H	H	4-Cl	Cl	O	121
54	H	CH ₃	H	H	H	4-Cl	OCF ₃	O	105
55	H	CH ₃	H	H	3-Cl	4-CN	Cl	O	140
56	H	CH ₃	H	H	3-Cl	4-CN	OCF ₃	O	98
57	H	H	OH	H	H	H	Cl	O	188
58	H	H	OH	H	H	H	OCF ₃	O	170
59	H	H	OH	H	H	4-Cl	Cl	O	Viscous
60	H	H	OH	H	H	4-Cl	OCF ₃	O	185
61	H	H	OH	H	H	4-Cl	OCF ₃	O	E-form 95
62	H	H	OH	H	H	4-CN	Cl	O	Z-form Viscous
63	H	H	OH	H	H	4-CN	OCF ₃	O	113
64	H	H	CH ₃	H	H	H	Cl	O	164
65	H	H	CH ₃	H	H	H	OCF ₃	S	118

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp □
66	H	H	OCH ₃	H	H	H	Cl	0	183
67	H	H	OCH ₃	H	H	H	OCF ₃	0	181
68	H	H	OC ₃ H ₇ -i	H	H	H	Cl	0	155
69	H	H	OC ₃ H ₇ -i	H	H	H	OCF ₃	0	193
70	H	H	OC ₄ H ₉ -i	H	H	H	Cl	0	176
71	H	H	OC ₄ H ₉ -i	H	H	H	OCF ₃	0	184
72	H	H	O-CO-CH ₃	H	H	H	OCF ₃	0	182
73	H	H	O-CO-Ph	H	H	H	OCF ₃	0	168
74	H	H	OH	CH ₃	H	H	Cl	0	115
75	H	H	OH	CH ₃	H	H	OCF ₃	0	130
76	H	H	H	H	3-F	4-CN	SCF ₃	0	214
77	H	H	H	H	3-F	4-CN	SOCF ₃	0	214
78	H	H	H	H	4-F	4-CN	SO ₂ CF ₃	0	165
79	H	H	H	H	3-Cl	4-CN	SOCF ₃	0	157

Table 1 (Cont'd)

No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	W	mp °
80	H	H	H	H	3-CF ₃	4-CN	SCF ₃	O	215
81	H	H	H	H	3-CF ₃	4-CN	SO ₂ CF ₃	O	210
82	H	H	H	H	3-CF ₃	4-CN	OCF ₃	O	152
83	H	H	H	H	3-CF ₃	4-CN	Cl	O	Z-form 165

Note: Ph is phenyl group.

Formula (I-2)

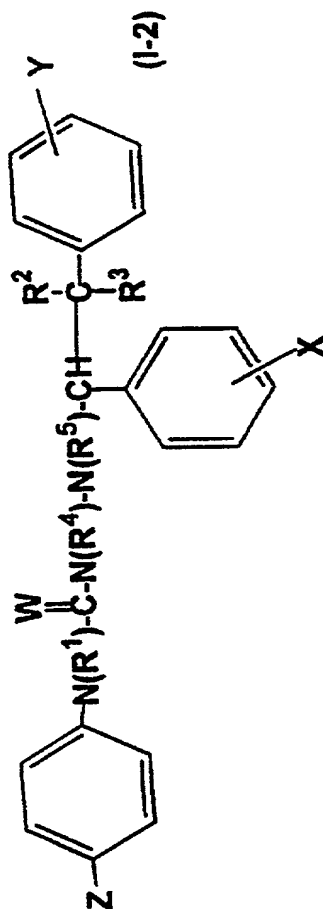


Table 2 (R¹ and R³ are hydrogen atoms)

No.	R ²	R ⁴	R ⁵	X	Y	Z	W	mp (°C)
84	H	H	H	H	H	Cl	O	211
85	H	H	H	H	H	OCF ₃	O	194
86	H	H	H	H	4-Cl	OCF ₃	O	209
87	H	H	H	H	4-CN	OCF ₃	O	204
88	H	H	H	H	4-NO ₂	OCF ₃	O	203
89	H	H	H	3-F	4-Cl	OCF ₃	O	203
90	H	H	H	3-Cl	4-Cl	OCF ₃	O	176

Table 2 (Cont'd)

No.	R ²	R ⁴	R ⁵	X	Y	Z	W	mp °
91	H	H	H	3-Cl	4-CN	OCF ₃	O	193
92	H	H	H	3-Cl	4-CN	SCE ₃	O	177
93	H	H	H	3-Cl	4-CN	SOCF ₃	O	178
94	H	H	H	3-Cl	4-CN	SO ₂ CF ₃	O	170
95	H	H	H	3-Br	4-CN	OCF ₃	O	187
96	H	H	H	3-CF ₃	4-CN	OCF ₃	O	165
97	H	H	H	3-CF ₃	4-CN	SCE ₃	O	164
98	H	H	H	H	4-Cl	OCF ₃	S	171
99	H	H	H	3-Cl	4-CN	OCF ₃	S	149
100	H	H	H	3-CF ₃	4-CN	OCF ₃	S	209
101	H	H	CO-CH ₃	3-Cl	4-CN	OCF ₃	O	178
102	H	H	CO-Ph	3-Cl	4-CN	OCF ₃	O	221

Table 2 (Cont'd)

No.	R ²	R ⁴	R ⁵	X	Y	Z	W	mp (J)
103	H	H	CONHC ₂ H ₅	3-Cl	4-CN	OCF ₃	O	201
104	H	OH	H	H	H	OCF ₃	O	190
105	H	OCH ₃	H	H	H	Cl	O	195
106	H	OCH ₃	H	H	H	OCF ₃	O	183
107	H	OCH ₃	H	H	H	OCF ₃	O	186
108	CH ₃	H	H	3-Cl	4-CN	OCF ₃	O	156
109	H	H	H	H	4-F	OCF ₃	O	209
110	H	H	H	H	4-Br	Cl	O	233
111	H	H	H	H	4-Br	OCF ₃	O	201
112	H	H	H	H	3-CN	OCF ₃	O	176
113	H	H	H	H	2-NO ₂	OCF ₃	O	197
114	H	H	H	3-F	4-CN	OCF ₃	O	189

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Table 2 (Cont'd)

No.	R ²	R ⁴	R ⁵	X	Y	Z	W	mp [°C]
115	H	H	H	3-F	4-CN	SCF ₃	O	189
116	H	H	H	3-F	4-CN	SO ₂ CF ₃	O	166
117	H	H	H	3-CF ₃	4-CN	OCF ₃	O	131
								(-) - Isomer
118	H	H	H	3-CF ₃	4-CN	OCF ₃	O	126
								(+) - Isomer
119	H	H	H	3-CF ₃	4-CN	SO ₂ CF ₃	O	Glassy
120	H	H	H	3-CF ₃	4-CN	SO ₂ CF ₃	O	Glassy
121	H	H	H	H	3-CN	OCF ₃	O	120

Note: Ph is phenyl group.

Compounds 106 and 107 are diastereomers.

Compound 106 is higher than Compound 107 in the R_f value.

5 Formula (I-3)

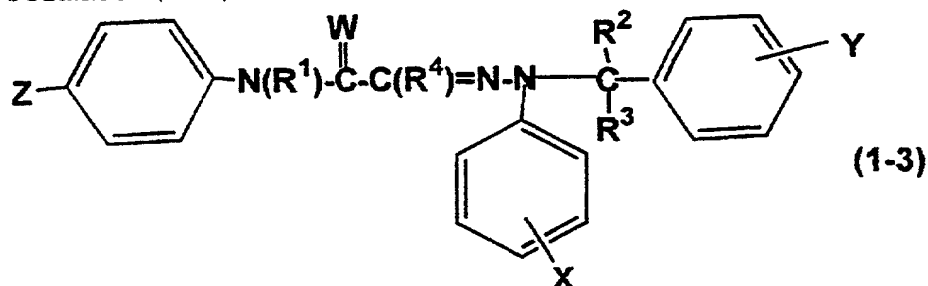


Table 3 (R^2 and R^3 are hydrogen atoms, and W is oxygen atom.)

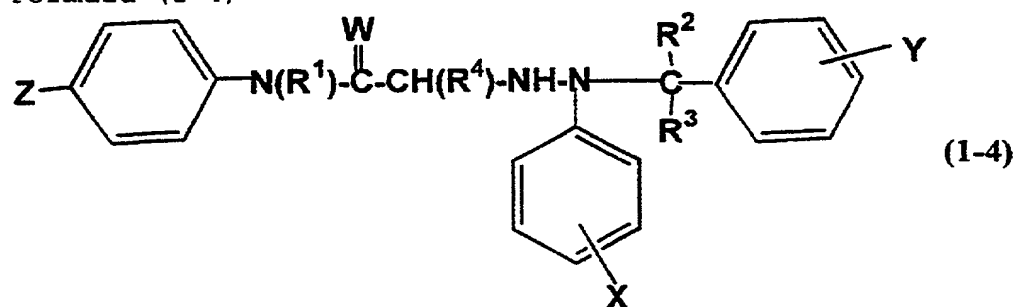
No	R^1	R^2	X	Y	Z	mp \square , Refractive index
122	H	H	H	H	OCF ₃	113.3-114.0
123	H	H	H	4-Cl	OCF ₃	137.8
124	H	H	H	4-CN	Cl	163
125	H	H	H	4-CN	OCF ₃	138
126	H	H	3-Cl	4-Cl	Cl	143.5-144.0
127	H	H	3-Cl	4-Cl	OCF ₃	139.6-141.5
128	H	H	3-Cl	4-NO ₂	Cl	174.0-176.5
129	H	H	3-Cl	4-NO ₂	OCF ₃	151.6-151.7
130	H	H	3-Cl	4-CN	Cl	191.0-192.0
131	H	H	3-Cl	4-CN	OCF ₃	160.5-162.0
132	H	H	3-Cl	4-CN	SCF ₃	188.0
133	H	H	3-Cl	4-CN	SOCF ₃	206.1
134	H	H	3-F	4-CN	Cl	154-156
135	H	H	3-F	4-CN	OCF ₃	155.9-156.8

Table 3 (Cont'd)

No	R ¹	R ⁴	X	Y	Z	mp °C, Refractive index
136	H	H	3-CH ₃	4-CN	Cl	127
137	H	H	3-CH ₃	4-CN	OCF ₃	166
138	H	H	3-CF ₃	4-CN	Cl	164-165
139	H	H	3-CF ₃	4-CN	OCF ₃	151.0
140	H	CH ₃	3-Cl	4-CN	OCF ₃	nD 1.5950 (25°C)
141	CH ₃	H	3-CF ₃	4-CN	Cl	209-211
142	H	H	3-Cl	2-CN	OCF ₃	148

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Formula (I-4)

Table 4 (R^1 , R^2 , R^3 and R^4 are hydrogen atoms.)

No	X	Y	Z	mp \square , Refractive index
143	H	H	OCF ₃	51.0-53.0
144	H	4-Cl	OCF ₃	92.1
145	H	4-CN	Cl	106-108
146	H	4-CN	OCF ₃	nD 1.5685 (27 \square)
147	3-Cl	4-Cl	Cl	105.3-106.4
148	3-Cl	4-Cl	OCF ₃	38.0
149	3-Cl	4-NO ₂	Cl	Viscous
150	3-Cl	4-NO ₂	OCF ₃	Viscous
151	3-Cl	4-CN	Cl	153.1
152	3-Cl	4-CN	OCF ₃	43.5-45.0
153	3-F	4-CN	Cl	164-165
154	3-F	4-CN	OCF ₃	nD 1.5615 (27 \square)
155	3-CH ₃	4-CN	Cl	138-139
156	3-CH ₃	4-CN	OCF ₃	nD 1.5315 (28 \square)
157	3-CF ₃	4-CN	Cl	43
158	3-CF ₃	4-CN	OCF ₃	153.1

Some of the compounds shown in Tables 1 to 4 are viscous or glassy substances. Their ^1H -NMR data are summarized in Table 5.

Table 5

No	^1H -NMR[CDCl_3/TMS , δ (ppm)]
59	6.29 (s, 1H), 7.65-7.92 (m, 13H), 9.14 (bs, 1H), 10.70 (bs, 1H). (DMSO- d_6)
62	3.88 (bs, 1H), 3.87 (s, 1H), 6.91-7.55 (m, 13H), 7.73 (s, 1H), 8.13 (bs, 1H).
119	3.12 (dd, 1H), 3.23 (dd, 1H), 4.12-4.32 (m, 2H), 6.13 (bs, 1H), 7.24-7.93 (m, 12H), 8.08 (bs, 1H).
120	3.11 (dd, 1H), 3.23 (dd, 1H), 4.13-4.28 (m, 2H), 5.97 (s, 1H), 7.25-7.75 (m, 12H), 7.90-8.00 (bs, 1H).
149	3.65 (d, 2H), 4.20 (t, 1H), 4.70 (s, 2H), 6.85 (dd, 1H), 6.93 (dd, 1H), 7.08 (dd, 1H), 7.15-7.21 (m, 3H), 7.24 (d, 2H), 7.40 (d, 2H), 8.13 (d, 2H), 8.40 (s, 1H).
150	3.64 (s, 2H), 4.69 (s, 2H), 6.84 (dd, 1H), 6.94 (dd, 1H), 7.09 (m, 3H), 7.23 (t, 1H), 7.29 (d, 2H), 7.40 (d, 2H), 8.12 (d, 2H), 8.40 (s, 1H).

5 The ant controller of the present invention
exhibits a markedly high killing effect at a low dosage
upon all the termites doing harm to houses, construction
materials, furniture, leathers, fibers, vinyl articles,
electric wires and cables, for example, RHINOTERMITIDAE
10 including *Coptotermes formosanus* Shiraki, *Reticulitermes*
speratus (Kolbe), *Reticulitermes hesperus* which inhabits
the North America, *Reticulitermes tibialis*,
Reticulitermes flavipes, *Reticulitermes lucifugus* which
inhabits the shore of the Mediterranean, *Reticulitermes*
15 *santonensis*, *Incisitermes minor* (Hagen), TERMITIDAE
including *Odontotermes formosanus* (Shiraki),
KALOTERMITIDAE including *Cryptotermes domesticus*
(Haviland), TERMOPSIDAE including *Hodotermopsis japonica*
(Holmgren), etc.

20 Further, the ant controller of the present
invention exhibits a markedly high killing effect at a
low dosage upon all the ants doing harm to crops, or to
human being when the ants invade into houses and public
facilities such as parks, for example, FORMICIDAE
25 including *Monomorium pharaonis* Linne, *Monomorium*
nipponense Wheellex, *Camponotus kiusiuensis* Santschi,
Formica japonica Motschulsky, *Lasius fuliginosus*
(Latreille), *Solenopsis richteri*, *Solenopsis invicta*,
Solenopsis geminata (Fireant), etc.

30 For using the ant controller of the present
invention containing the hydrazine derivative of formula
(I) as an active ingredient efficiently, the ant

As the solid carrier used in the present invention, there can be exemplified clays such as kaolin, bentonite, acid clay and the like; talcs such as talc, pyrophyllite and the like; silica materials such as diatomaceous earth, siliceous sand, mica, synthetic silicate, synthetic high-dispersion silica and the like; and inorganic mineral powders such as pumice, sand and the like; organic matters such as pieces of wood, chips of pulp wood, grain flour, sugars and the like. As the liquid carrier, there can be exemplified alcohols such as methyl alcohol, ethyl alcohol, ethylene glycol and the like; ketones such as acetone, methyl ethyl ketone, cyclohexanone and the like; ethers such as ethyl ether, dioxane, tetrahydrofuran, Cellosolves and the like; aliphatic hydrocarbons such as light oil, kerosene and

As the solid carrier used in the present invention, there can be exemplified clays such as kaolin, bentonite, acid clay and the like; talcs such as talc, pyrophyllite and the like; silica materials such as diatomaceous earth, siliceous sand, mica, synthetic silicate, synthetic high-dispersion silica and the like; and inorganic mineral powders such as pumice, sand and the like; organic matters such as pieces of wood, chips of pulp wood, grain flour, sugars and the like. As the liquid carrier, there can be exemplified alcohols such as methyl alcohol, ethyl alcohol, ethylene glycol and the like; ketones such as acetone, methyl ethyl ketone, cyclohexanone and the like; ethers such as ethyl ether, dioxane, tetrahydrofuran, Cellosolves and the like; aliphatic hydrocarbons such as light oil, kerosene and

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5 the like; aromatic hydrocarbons such as benzene,
toluene, xylene, solvent naphtha, cyclohexanone,
methylnaphthalene and the like; and halogenated
hydrocarbons such as chloroform, carbon tetrachloride,
chlorobenzene and the like. These solid and liquid
10 carriers may be used either alone or in the form of a
mixture.

As the auxiliaries which can be used in the
present invention, surfactants, dispersants, sticking
agents, etc. can be referred to. As the surfactants,
15 there can be exemplified polyoxyethylene alkylaryl
ethers, polyoxyethylene sorbitan monolaurates, alkylaryl
sorbitan monolaurates, alkylbenzenesulfonates,
alkylnaphthalene-sulfonates, ligninsulfonates, higher
alcohol sulfuric ester salts, etc. These surfactants
20 may be used either alone or in the form of a mixture.

As the dispersants or sticking agents, for
example, casein, gelatin, starch, alginic acid,
carboxymethyl cellulose, agar, polyvinyl alcohol,
turpentine oil, etc. can be used according to the need.

25 The ant controller of the present invention is
applied not only to the surrounding soil surface or into
the under-floor soil in order to protect wooden
materials such as trees, board fences, sleepers, etc.
and structures such as shrines, temples, houses,
30 outhouses, factories, etc., but it can also be applied
to lumbered articles such as surfaces of the under-floor
concrete, alcove posts, beams, plywoods, furniture,

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5 etc., wooden articles such as particle boards, half
boards, etc. and vinyl articles such as coated electric
wires, vinyl sheets, heat insulating material such as
styrene foams, etc. In case of application against ants
doing harm to crops or human beings, the ant controller
10 of the present invention is applied to the crops or the
surrounding soil, or is directly applied to the nest of
ants or the like.

The present invention is not limited to the
embodiments mentioned above, but it also includes the
15 embodiments of applying the ant controller of the
invention preventively to places at which occurrence of
ants is expected.

In putting the ant controller of the present
invention, the dosage may be appropriately selected from
20 the ranges properly chosen. In case of application to
wooden materials, the quantity of active ingredient
ranges from 0.1 to 50 g per m²; and in case of soil
treatment or application to the nests, the quantity of
active ingredient ranges from 1 to 500 g per m².

25 EXAMPLES

Next, typical examples and test example of the
present invention are presented below. The invention is
by no means limited to these examples.

In the examples, "parts" are by weight.

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5 Formulation Example 1

Each hydrazine derivative listed

in Tables 1-4 20 parts

Xylene 80 parts

10 The ingredients mentioned above were made into
a uniform solution to obtain an oily solution.

Formulation Example 2

Each hydrazine derivative listed

in Tables 1-4 10 parts

Polyoxyethylene styrylphenyl ether 10 parts

15 Cyclohexanone 80 parts

The ingredients mentioned above were uniformly
mixed and dissolved together to obtain an emulsifiable
concentrate.

Formulation Example 3

20 Each hydrazine derivative listed

in Tables 1-4 10 parts

Sodium alkylbenzenesulfonate 2 parts

White carbon 10 parts

Clay 78 parts

25 The ingredients mentioned above were uniformly
mixed and pulverized to obtain a wettable powder.

Formulation Example 4

Each hydrazine derivative listed

in Tables 1-4 8 parts

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Table 6

Compound No.	Termite-killing effect	Compound No.	Termite-killing effect
1	A	5	A
2	B	6	A
3	A	7	A
4	A	8	C

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5 Table 6 (Cont'd)

Compound No.	Termite-killing effect	Compound No.	Termite-killing effect
9	B	32	A
10	A	33	C
11	A	34	A
12	A	35	A
13	A	36	B
14	A	37	A
15	B	38	B
16	C	39	A
17	A	40	D
18	A	41	A
19	A	42	A
20	A	43	A
21	A	44	C
22	B	45	A
23	A	46	A
24	C	47	A
25	D	48	A
26	A	49	C
27	A	50	A
28	C	51	A
29	C	52	A
30	A	53	B
31	A	54	A

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5 Table 6 (Cont'd)

Compound No.	Termite-killing effect	Compound No.	Termite-killing effect
55	A	78	A
56	A	79	B
57	D	80	A
58	A	81	A
59	C	82	B
60	C	83	D
61	A	84	A
62	A	85	C
63	A	86	A
64	A	87	C
65	C	88	A
66	A	89	B
67	A	90	A
68	A	91	A
69	B	92	A
70	A	93	D
71	A	94	A
72	A	95	A
73	A	96	A
74	A	97	A
75	A	98	A
76	A	99	A
77	A	100	A

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5 Table 6 (Cont'd)

Compound No.	Termite-killing effect	Compound No.	Termite-killing effect
101	A	124	D
102	A	125	A
103	A	126	A
104	A	127	A
105	B	128	A
106	A	129	A
107	D	130	C
108	C	131	C
109	C	132	A
110	B	133	A
111	D	134	A
112	A	135	B
113	A	136	A
114	B	137	A
115	A	138	A
116	B	139	A
117	A	140	A
118	D	141	D
119	A	142	C
120	A	143	C
121	C	144	B
122	D	145	A
123	A	146	D

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5 Table 6 (Cont'd)

Compound No.	Termite-killing effect	Compound No.	Termite-killing effect
147	A	153	A
148	A	154	B
149	A	155	A
150	C	156	B
151	C	157	A
152	B	158	C

Test Example 2

10 The ant controller of the present invention was applied to nests (anthill) of fireant (*Solenopsis geminata*) with drench treatment, in terms of 1 g of the active ingredient per one nest. 14 Days after the treatment of the ant controller, the activity of the nests was evaluated according to the following

15 criterion. The test was carried out with one block per one nest.

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5	Criterion	Effect
	A	Nest is completely destructed or activity of the nest is extremely low.
	B	Activity of the nest is exhibited.
	C	High activity of the nest is exhibited.
10	D	Activity of the nest is extremely high.

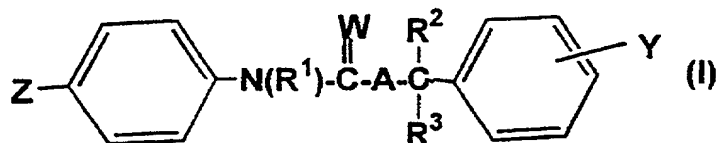
As a result of the test, compound Nos. 44 and 96 of the present invention exhibited the effect "A".

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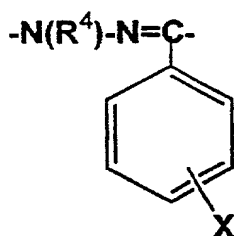
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WHAT IS CLAIMED IS:

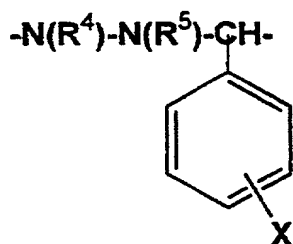
1. An ant controller characterized by containing, as active ingredient thereof, a hydrazine derivative represented by the following formula (I):



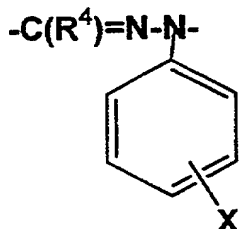
wherein A represents:



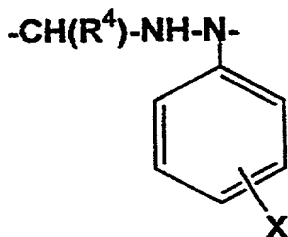
(wherein R^4 represents hydrogen atom or C_1 - C_6 alkyl group, and X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C_1 - C_6 alkyl group and halo C_1 - C_6 alkyl group),



(wherein R^4 and X are as defined above, and R^5 represents hydrogen atom, C_1 - C_6 alkylcarbonyl group or phenylcarbonyl group which may have 1 to 2, same or different substituents selected from the group consisting of C_1 - C_6 alkyl groups),



(wherein R^4 and X are as defined above), or



(wherein R^4 and X are as defined above);

R^1 represents hydrogen atom or C_1 - C_6 alkyl group;

R^2 and R^3 , which may be same or different, represent hydrogen atom, hydroxyl group, C_1 - C_6 alkyl group, C_1 - C_6 alkoxy group, C_1 - C_6 alkylcarbonyl group or phenylcarbonyl group;

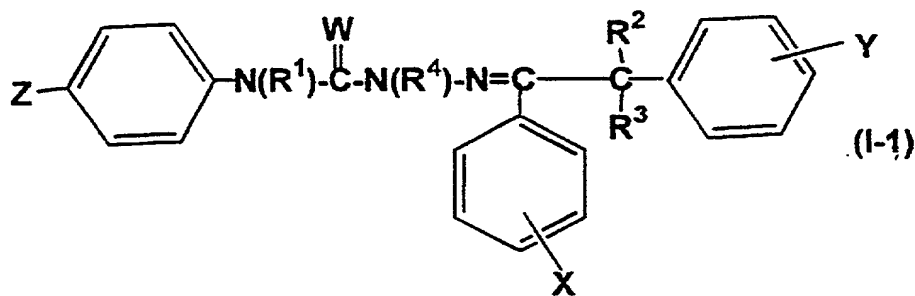
Y represents 1 to 5, same or different substituents selected from the group consisting of

hydrogen atom, halogen atom, nitro group and cyano group;

Z represents halogen atom, cyano group, C₁-C₆ alkyl group, halo C₁-C₆ alkyl group, C₁-C₆ alkoxy group, halo C₁-C₆ alkoxy group, halo C₁-C₆ alkylthio group, halo C₁-C₆ alkylsulfinyl group or halo C₁-C₆ alkylsulfonyl group; and

W represents oxygen atom or sulfur atom.

2. The ant controller according to Claim 1, which is represented by the following formula (I-1):



wherein R¹ represents hydrogen atom or C₁-C₆ alkyl group;

R² and R³, which may be same or different, represent hydrogen atom, hydroxyl group, C₁-C₆ alkyl group, C₁-C₆ alkoxy group, C₁-C₆ alkylcarbonyl group or phenylcarbonyl group;

R⁴ represents hydrogen atom or C₁-C₆ alkyl group;

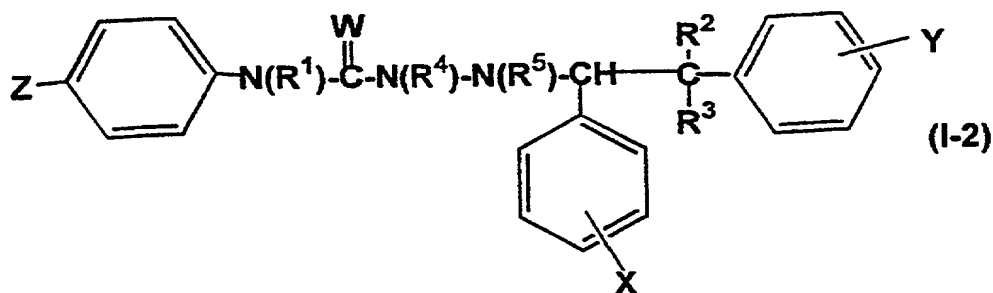
X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C₁-C₆ alkyl group and halo C₁-C₆ alkyl group;

Y represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, nitro group and cyano group;

Z represents halogen atom, cyano group, C₁-C₆ alkyl group, halo C₁-C₆ alkyl group, C₁-C₆ alkoxy group, halo C₁-C₆ alkoxy group, halo C₁-C₆ alkylthio group, halo C₁-C₆ alkylsulfinyl group or halo C₁-C₆ alkylsulfonyl group; and

W represents oxygen atom or sulfur atom.

3. The ant controller according to Claim 1, which is represented by the following formula (I-2):



wherein R¹ represents hydrogen atom or C₁-C₆ alkyl group;

R² and R³, which may be same or different, represent hydrogen atom, hydroxyl group, C₁-C₆ alkyl group, C₁-C₆ alkoxy group, C₁-C₆ alkylcarbonyl group or phenylcarbonyl group;

R⁴ represents hydrogen atom or C₁-C₆ alkyl group;

R⁵ represents hydrogen atom, C₁-C₆ alkylcarbonyl group or phenylcarbonyl group which may

have 1 to 2, same or different substituents selected from the group consisting of C₁-C₆ alkyl groups;

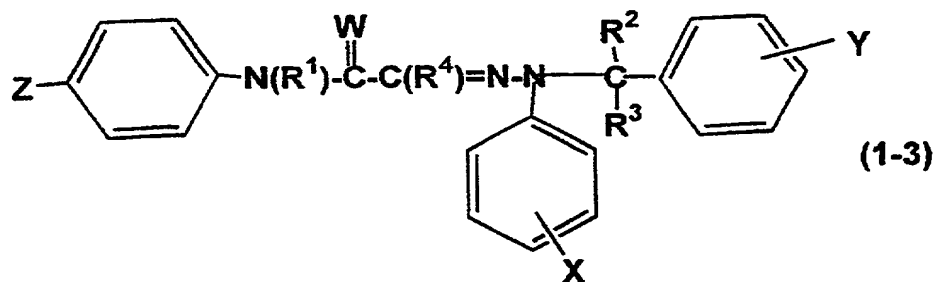
X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C₁-C₆ alkyl group and halo C₁-C₆ alkyl group;

Y represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, nitro group and cyano group;

Z represents halogen atom, cyano group, C₁-C₆ alkyl group, halo C₁-C₆ alkyl group, C₁-C₆ alkoxy group, halo C₁-C₆ alkoxy group, halo C₁-C₆ alkylthio group, halo C₁-C₆ alkylsulfinyl group or halo C₁-C₆ alkylsulfonyl group; and

W represents oxygen atom or sulfur atom.

4. The ant controller according to Claim 1, which is represented by the following formula (I-3):



wherein R¹ represents hydrogen atom or C₁-C₆ alkyl group;

R² and R³, which may be same or different, represent hydrogen atom, hydroxyl group, C₁-C₆ alkyl group, C₁-C₆ alkoxy group, C₁-C₆ alkylcarbonyl group or

phenylcarbonyl group;

R^4 represents hydrogen atom or C_1-C_6 alkyl group;

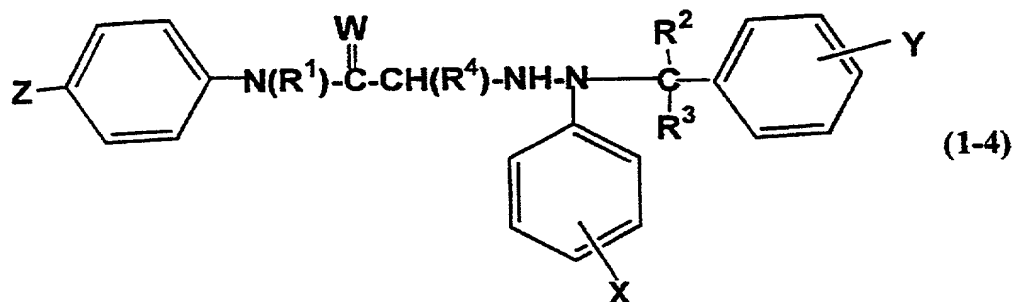
X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C_1-C_6 alkyl group and halo C_1-C_6 alkyl group;

Y represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, nitro group and cyano group;

Z represents halogen atom, cyano group, C_1-C_6 alkyl group, halo C_1-C_6 alkyl group, C_1-C_6 alkoxy group, halo C_1-C_6 alkoxy group, halo C_1-C_6 alkylthio group, halo C_1-C_6 alkylsulfinyl group or halo C_1-C_6 alkylsulfonyl group; and

W represents oxygen atom or sulfur atom.

5. The ant controller according to Claim 1, which is represented by the following formula (I-4):



wherein R^2 represents hydrogen atom or C_1-C_6 alkyl group;

R^2 and R^3 , which may be same or different,

represent hydrogen atom, hydroxyl group, C₁-C₆ alkyl group, C₁-C₆ alkoxy group, C₁-C₆ alkylcarbonyl group or phenylcarbonyl group;

R⁴ represents hydrogen atom or C₁-C₆ alkyl group;

X represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, C₁-C₆ alkyl group and halo C₁-C₆ alkyl group;

Y represents 1 to 5, same or different substituents selected from the group consisting of hydrogen atom, halogen atom, nitro group and cyano group;

Z represents halogen atom, cyano group, C₁-C₆ alkyl group, halo C₁-C₆ alkyl group, C₁-C₆ alkoxy group, halo C₁-C₆ alkoxy group, halo C₁-C₆ alkylthio group, halo C₁-C₆ alkylsulfinyl group or halo C₁-C₆ alkylsulfonyl group; and

W represents oxygen atom or sulfur atom.

6. A method for application of an ant controller which comprises treating a wooden part and a surrounding soil where ants and termites live, with an effective quantity of the ant controller according to Claim 1.

7. The method for application of an ant controller according to Claim 6, wherein the hydrazine derivative represented by the general formula (I) is a hydrazine derivative claimed in any one of Claims 2 to 5.

2040407 T846T001

**CLAIM FOR BENEFIT OF EARLIER U. S. / PCT APPLICATION(S)
UNDER 35 U. S. C. 120**

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that / those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national of PCT international filing date of this application.

U. S. Application(s) (or PCT applications designating U. S.)

U. S. Application Serial No.	Filing Date	Status (pending, patented, abandoned)

RELATED FOREIGN APPLICATIONS

Related foreign applications, if any, filed in the name of the inventor(s) or the inventor(s) assigns more than twelve months before the filing of the subject application are as follows

Country	Application No.	Date of filing	Date of issue or publication

AM100,246

DECLARATION AND POWER OF ATTORNEY AND PETITION

As a below named inventor, I hereby declare that:

INVENTORSHIP IDENTIFICATION

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ANT CONTROLLERS AND METHOD FOR APPLICATION THEREOF

SPECIFICATION IDENTIFICATION

the specification of which

☐ is attached hereto.

☐ was filed on _____ as

Application Serial No. _____

and was amended on _____ (if applicable).

☒ was filed as PCT international application

Number _____ PCT/US 00/17895

on _____ June 28, 2000 ,

and was amended under PCT Article 19

on _____ (if applicable)

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information known by me to be material to the patentability of this application in accordance with Title 37, Code of the Federal Regulations. §1.56(a).

☐ In compliance with this duty there is attached an information disclosure statement. 37 CFR 1.97.

☒ In compliance with this duty, information which may be material is disclosed in the specification of the subject application.

POWER OF ATTORNEY

I hereby appoint the following attorney(s) and agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected herewith:

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 Mary K. Cameron
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 Reg. No. 36,439
 Reg. No. 36,814
 Reg. No. 31,278
 Reg. No. 42,906
 Reg. No. 30,977
 Reg. No. 42,220
 Reg. No. 39,150
 Reg. No. 43,118
 Reg. No. 45,639
 Reg. No. 46,295

14

each of the above attorney(s) and agent(s) to have full powers of substitution and revocation, and each and any attorney or agent so substituted to have full powers of substitution and revocation.

Address all telephone calls to: at telephone no. (973) 426-

Address all correspondence to: BASF Corporation, Patent Department; 3000 Continental Drive-North,
 Mount Olive, NJ 07828-1234

CLAIM FOR BENEFIT OF FOREIGN PRIORITY UNDER 35 U. S. C. §119

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United State of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

- ☐ No such applications have been filed.
☒ Such applications have been filed as follows

**DETAILS OF FOREIGN APPLICATION FROM WHICH PRIORITY CLAIMED
 UNDER 35 U. S. C. §119**

Country	Application No.	Date of filing	Date of issue or publication
Japan	H11(1999)-190671	05 July 1999	

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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